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METHODS FOR MEASURING THE INTENSITIES OF AITKEN NUCLEI SOURCES IN THE ATMOSPHERE

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METHODS FOR MEASURING THE INTENSITIES OF AITKEN NUCLEI SOURCES IN THE ATMOSPHERE

(Methodes de mesure de l'intensite des sources de noyaux d'aitken dans l'atmosphere)

A. Lopez, J. Servant
and J. Fontan

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METHODS FOR MEASURING THE INTENSITIES OF AITKEN NUCLEI SOURCES IN THE ATMOSPHERE

Application of Several of the Principal Sources in Southwestern France

A. Lopez*, J. Servant* and J. Fontan*

ABSTRACT. Different methods for measuring the intensities of Aitken nuclei sources in the atmosphere are presented. They are based on data obtained from aircraft measurements. The intensity of a small source area, some km^2, is obtained by the vertical repartition of the concentrations in the plume; for larger source area, some 10^4 km^2, this intensity is determined from the variation with day time of the vertical profiles of the concentration. The accuracy of the different methods is discussed.

The values obtained for some of the most intense sources of the southwestern part of France are given: the urban district of Toulouse with 400,000 inhabitants (flux: 5.10^{17} nuclei s^{-1}); the industrial site of Lacq (flux: 1.10^{18} nuclei s^{-1}); the coniferous forest of Landes (intensity: 1.10^5 nuclei cm^{-2} s^{-1}); the rural area of the

*Paul Sabatier University, Aerosol Atomic Physics and Atmospheric exchange center, Research Team associated with CNRS No. 378, 31077 Toulouse, Cedex, France.

Midi-Pyrenees (intensity: 3.10^4 nuclei cm^{-2} s^{-1}). From these results, it is deduced that the natural sources of the region are stronger than the artificial sources (75 percent compared with 25).

Introduction

In the atmosphere, there exist two types of Aitken nuclei sources; those of natural origin, linked with the transformation reactions of minor gaseous constituents arising from the soil or vegetation, and those of artificial origin, linked with industrial and domestic pollution. It is difficult to actually assess the relative importances of these different sources.

In this work, we present three methods for the determination of Aitken nuclei source intensity. They are based on the study of vertical repartitions of nuclei in the atmosphere. Each is elaborated according to different principles: determination of particle flux in a plume, evolution of the vertical repartitions of nuclei in an air mass as a function of time, or as a function of the distance the mass has traversed above a source. Each of them is specific for the study of a certain geometric type of source (source of reduced dimensions: urban centers and industrial complexes, for example, or a source of large surface area: zone of vegetation, for example).

These methods of measurement have been applied to several of the principal nuclei sources in southwestern France: the urban center of Toulouse (population 400,000), the industrial complex of Lacq, coniferous vegetation in the forest of Landes, and rural zones of the Midi-Pyrenees region. However fragmentary, these results permit us to make a first balance, and thus to obtain an estimate of the relative importances of natural and artificial sources in the region.

Experimental Methods

Measurements are made during periods of fair weather with the aid of a Cessna TU 206, equipped as follows:

For continuous recording of meteorological parameters; temperature, humidity, dew point. Direction and intensity of the wind are measured by Doppler radar.

For the measurement of nuclei concentrations: a counter, which has been described (Cabrol et al., 1972). The frequency of samples, one per minute) corresponds to a measurement integrated over a distance of about 2.5 km (speed of plane is approximately 150 km h^{-1}). In certain cases (plume studies, §I), we utilized a system of electrostatic precipitators designed to measure the concentrations of small positive and negative ions (Guedalia et al., 1972). This indirect method for determination of the number of Aitken nuclei presents the advantage of a time constant on the order of a second, corresponding to a resolution on the order of 40 meters. Considering that the average radius of particles in the plume does not vary, it may be shown that, at equilibrium, the small ion concentration (n) is inversely proportional to the Aitken nuclei concentration (N) (Guedalia et al., 1972).

$$n = a/N. \qquad (1)$$

The proportionality constant a depends on the intensity of ionization and on the dimension of the particles. It is determined experimentally for each sample level by comparing at one point simultaneously measured small ion and nuclei concentrations.

1. Nuclei Source Contributing to the Establishment of a Plume

Let us consider a nuclei source, situated near the ground, of constant intensity as a function of time. Diffusion and advection contribute to the establishment of a plume (Figure 1).

1.1. Principle. With the aid of an airplane, one determines the repartition of particle concentrations in a vertical section of the plume situated at x_1 from the zone of emission. These concentrations are obtained, in measurement levels at different altitudes, by cutting the polluted zone perpendicularly to the direction of the wind. This permits a reconstruction of the plume, and, by knowing the wind speed, determination of particle flux through the vertical section being considered. This flux depends on the intensity of the source, as well as on the formation and disappearance of nuclei within the plume.

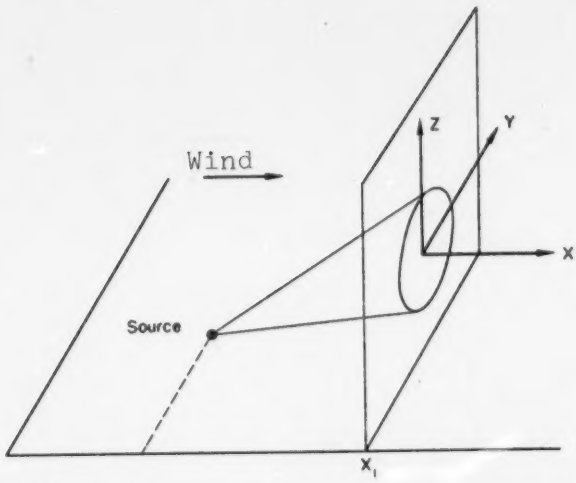

Figure 1. Schema of method for determining particle flux in a plume

The particle flux through a vertical section of the plume situated at distance x_1 from the source may be written:

$$\phi_{x_1} = \int_{-\infty}^{+\infty} \int_{0}^{+\infty} (C_{x_1}(y, z) - C_{x_{1,0}}(z)) V_{x_1}(z)\, dy\, dz. \qquad (2)$$

C_{x_1} (y, z) — volume concentration of nuclei in the plane x_1, inside the plume;

$C_{x_{1,0}}$ (z) — volume concentration of nuclei in the plane x_1, outside the plume. This value, corresponding to particle "background noise", is considered constant for a level of measurement;

V_{x_1} (z) — wind speed perpendicular to the plane x_1.

The integral is limited, on the y axis, by the width of the plume, and on the z axis, in general, during the day, by a stable stratum in the upper levels of the atmosphere.

The integral equation (2) transforms into a summation expression of the form:

$$\phi_{x_1} = \sum_{j=1}^{m} \sum_{i=1}^{n} (\overline{C_{x_1}}(i, j) - \overline{C_{x_{1,0}}}(j)) \overline{V_{x_1}}(j)\, \Delta y \,.\, \Delta z. \qquad (3)$$

Δy — distance between two measuring points on a sample level. This depends on the speed of the plane, and on the time constant for the measuring apparatus;

Δz — altitude between two levels of measurement, 150 m in general;

$\overline{C_{x_1}}(i,j)$ — mean volume concentration over the distance Δy, measured in the plume at a point having coordinates i (y axis), j (z axis);

$\overline{C_{x_{1,0}}}(j)$ — mean volume concentration outside the plume, on the measurement level of coordinate j;

$\overline{V_{x_1}}(j)$ — mean wind speed in the stratum j of thickness Δz.

Results obtained by the use of a summation formula will be increasingly precise as the number of measuring points within the plume becomes increasingly large. Along the y axis, this number depends on the width of the polluted zone, on the speed of the plane (with a reduced margin of variation), and on the time constant of the sampling apparatus. Along the z axis, the number of points will depend on the increment between two measurement levels.

1.1.1. Accuracy of the flux determination. An error calculation from Expression (3) gives:

$$\frac{\Delta\phi_{x_1}}{\phi_{x_1}} = \frac{\sum_{j=1}^{m}\left[\left\{\sum_{i=1}^{n}(\overline{C_{x_1}}(i,j)) - n\overline{C_{x_{1,0}}}(j)\right\}\Delta\overline{V_{x_1}}(j) + \overline{V_{x_1}}(j)\Delta\left\{\sum_{i=1}^{n}(\overline{C_{x_1}}(i,j)) - n\overline{C_{x_{1,0}}}(j)\right\}\right]}{\sum_{j=1}^{m}\left(\sum_{i=1}^{n}(\overline{C_{x_1}}(i,j)) - n\overline{C_{x_{1,0}}}(j)\right)V_{x_1}(j)} + \frac{\Delta(\Delta y)}{\Delta y} + \frac{\Delta(\Delta z)}{\Delta z}. \quad (4)$$

Considering a wind of constant intensity throughout the height of the polluted zone ($\overline{V_{x_1}}(j) = V$), and an absolute error in particle content closely proportional to the concentration (Table 1), this expression may be written, to a first approximation, in the form:

$$\frac{\Delta\phi_{x_1}}{\phi_{x_1}} = \frac{\Delta V}{V} + \frac{|\Delta C_{M_{x_1}}|}{C_{M_{x_1}} - C_{M_{x_{1,0}}}} + \frac{|\Delta C_{M_{x_{1,0}}}|}{C_{M_{x_1}} - C_{M_{x_{1,0}}}} + \frac{\Delta(\Delta y)}{\Delta y} + \frac{\Delta(\Delta z)}{\Delta z}. \quad (5)$$

$C_{M_{x_1}}$ — mean volume concentration in the plume;

$C_{M_{x_1,0}}$ — mean volume concentration exterior to the plume at a height corresponding to the thickness of the polluted zone.

The different experimental errors are as follows:

For wind intensity as given by Doppler radar installed in the plane, the relative error is on the order of 15 percent.

For the distance covered between two points of measurement, due essentially to variations in the speed of the plane, we estimate a 5 percent error.

For the altitude of the sample level, error is on the order of 5 percent.

TABLE 1. ABSOLUTE ERROR IN DETERMINATION OF NUCLEI CONCENTRATION

Absolute error (particles cm^{-3})	Measured concentration (particles cm^{-3})
500	10^3
750	10^4
1000	2.10^4
1250	3.10^4
1700	4.10^4
2000	6.10^4

For the determination of particle concentrations, the error is estimated through an imprecision of 0.5 percent in the extinction rate readings (Table 1).

In Expression (5) we may distinguish two terms: the first, linked with the airplane itself, contains in every case a relative error on the order of 25 percent; the second, dependent on measurements of particle concentration, contains progressively less error when the measured content is increasingly great.

One may notice, in the preceding calculations, that the geometric limits of the plume are assumed to be perfectly defined. In fact, when the concentrations within the plume are great in comparison with those outside the zone, in such a manner that

$$\Delta C_{M_{x_1}} + \Delta C_{M_{x_{1,0}}} \ll C_{M_{x_1}} - C_{M_{x_{1,0}}} , \qquad (6)$$

the supplementary error introduced by this hypothesis may be neglected in view of the preceding values.

On the other hand, when the interior concentrations are low, the above inequality is not verifiable. A supplementary source of error, significant but difficult to estimate, may alter preceding values. In such cases, precision of measurement is poor.

1.2. Particle flux issuing from the urban district of Toulouse. The city of Toulouse is situated 250 km from the Atlantic coast, 200 km from the Mediterranean coast, and 150 km north of the Pyrenees. There are 400,000 inhabitants within an area of 110 km^2. North-south, this area measures 15 km; east-west — 13 km; its perimeter, 71 km.

Figures 2 and 3 correspond to two characteristic examples obtained on November 26, 1971, and September 8, 1972, at a distance of 10 km from the center of the city. On November 26, 1971, we note a wind direction change below 600 m, provoking a displacement of the polluted zone toward the southwest. In both cases, a temperature inversion at 1200 m limits the altitude of the plume.

Figure 4 presents the results of a study executed on September 9, 1972, at 10 km from the center of the city. Concentrations of small ions and of nuclei are measured simultaneously.

The nuclei content, deduced from that of the small ions, is obtained through the intermediary of Equation (1). The proportionality constant is determined for each sample level from values for small ions and nuclei measured simultaneously, exterior to the plume.

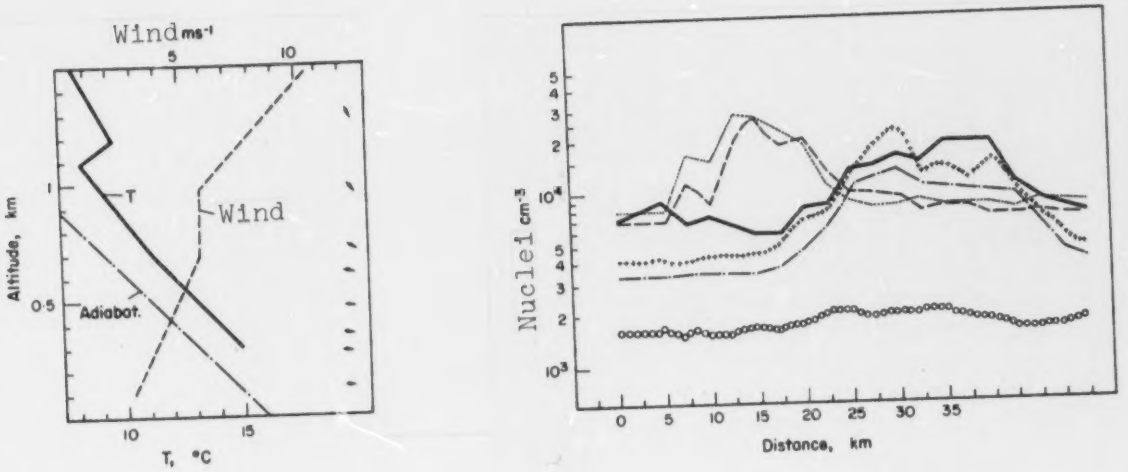

Figure 2. Experiment on November 26, 1971, in the plume from the urban district of Toulouse at 10 km from the center of the city. Measurement altitude at 300 m (-----) 450 m (·····); 600 m (——); 750 m (++++++); 1000 m (-·-·-); 1200 m (0000)

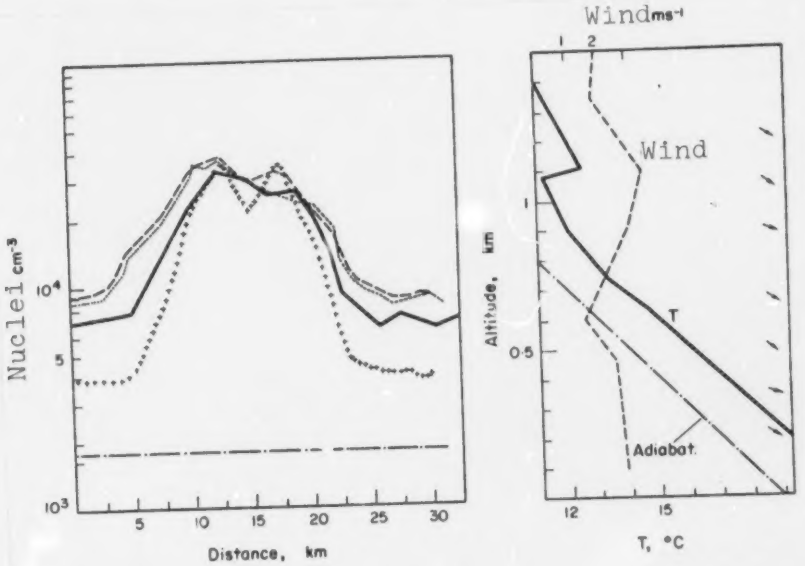

Figure 3. Experiment on September 8, 1972, in the plume from the urban district of Toulouse at 10 km from the center of the city. Measurement altitude at 150 m (-----); 300 m (·····); 600 m (——); 900 m (++++++); 1200 m (-·-·-)

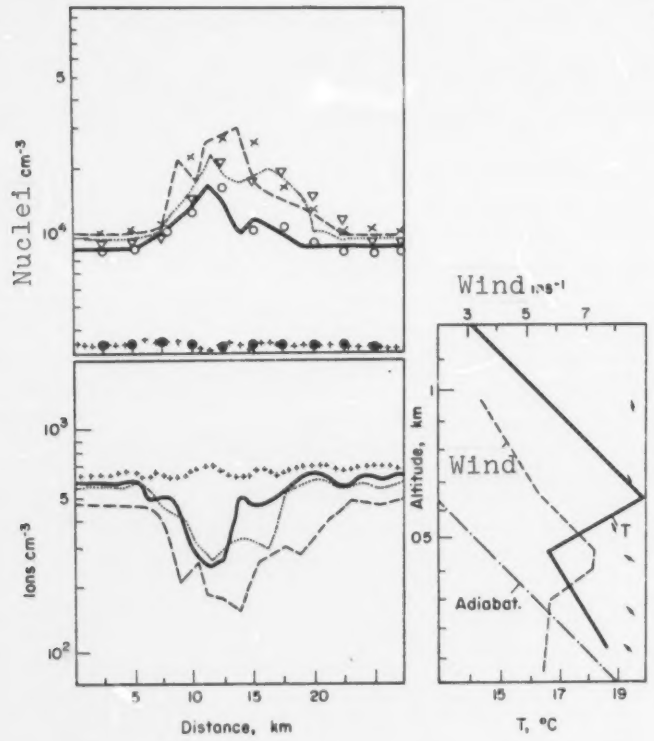

Figure 4. Experiment on September 9, 1972 in the plume from the urban district of Toulouse at 10 km from the center of the city. Measurement altitudes for small ion concentrations and calculated values for nuclei concentrations: 150 m (-----); 240 m (.....) 300 m (——); 450 m (++++++); direct measurement of nuclei concentrations: 150 m (×); 240 m (∇) 300 m (o); 450 m (●)

As we have already mentioned, the time constant (on the order of one second) for the experimental small ion measuring device permits a continuous tracing of the repartition of nuclei concentrations within the polluted zone. We note an adequate correlation to content values obtained with the aid of the particle counter, representing integrations over a distance of 2.5 km. The proportionality constant between concentrations of small ions and nuclei thus remains effectively the same inside and outside the plume. This implies a mean granulometry of the particles and a fairly stable ionization intensity in the air. It also implies that small ion

concentrations are very near to equilibrium values, which is reasonable, since the lifetime of these particles is on the order of 60 s for a 20,000 cm^{-3} concentration of nuclei with mean radius $3 \cdot 10^{-6}$ cm.

Six such measurements were made at 10 and 30 km from the city. For each case we have given (Table 2) flux values along with their corresponding precisions.

The mean flux value is on the order of $5 \cdot 10^{17}$ particles s^{-1}. The minimum value ($2.5 \cdot 10^{17}$ particles s^{-1}) is accompanied by a great imprecision. In effect, the mean concentrations inside and outside the polluted zone, 13,000 and 10,000 nuclei cm^{-3}, respectively, are such that Inequality (6) (see 1.1) is not verified. This leads to poor definition of the geometric limits of the plume, introducing, as mentioned previously, an error greater than that given by Equation (5). These geometric limits include, in the case under consideration, the width and height of the polluted zone (we observed no temperature inversion with altitude).

The relative error in flux determination for our experimental conditions, with the exception of the above-mentioned case, is between 35 and 50 percent. As we remarked earlier, the error inherent in the plane is 25%. Error in measurement of particle content varies between 10 and 25 per cent. This error is more significant when the flux value is low and when dilution of the polluted zone is great.

1.3. Particle flux issuing from the Lacq industrial complex. Situated near the urban district of Pau, the Lacq complex is comprised of industrial activities and natural gas treatment facilities.

Two characteristic examples of measurements made 5 km from the center are given in Figures 5 and 6.

In the case of Figure 6, the small ion and nuclei concentrations are measured simultaneously. As we mentioned concerning

TABLE 2. AITKEN NUCLEI FLUX EMITTED BY THE URBAN DISTRICT OF TOULOUSE

Date and time	Distance from city (km)	Polluted zone width* (km)	Mean wind speed (ms^{-1})	Particle flux (particles s^{-1})	Observations
November 26, 1971; 11:00 h	10	20	3	$7{,}0.10^{17} \pm 2{,}4.10^{17}$	Friday. Plume limited in altitude by a temperature inversion situated at 1200 m
April 25, 1972 10:00 h	10	20	13	$5{,}8.10^{17} \pm 2{,}9.10^{17}$	Tuesday. Plume limited in altitude by temperature inversion situated at 900 m
April 25, 1972 11:00 h	30	22	13	$5{,}4.10^{17} \pm 2{,}7.10^{17}$	Conditions identical to those of preceding measurement
April 26, 1972 11:00 h	30	25	3	$2{,}5.10^{17} \pm 1{,}6.10^{17}$	Wednesday. No temperature inversion with altitude — plume detected to height of 800 m
September 8, 1972; 14:00 h	10	18	3	$6{,}2.10^{17} \pm 2{,}5.10^{17}$	Friday. Plume limited in altitude by temperature inversion at 1200 m
September 9, 1972; 09:00 h	10	16	6	$3{,}5.10^{17} \pm 1{,}4.10^{17}$	Saturday. Plume limited in altitude by temperature inversion at 450 m

* The width of the polluted zone corresponds to the distance over which the plume is detected.

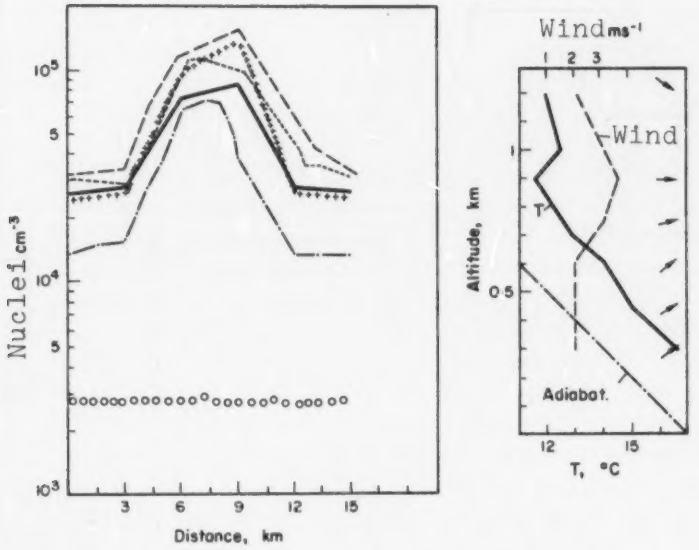

Figure 5. Experiment on June 21, 1972, in the plume from the Lacq industrial complex at 5 km from the center of emission. Measurement altitudes: 300 m (----); 450 m (·····); 600 m (——); 750 m (+++++++); 900 m (-·-·-); 1200 m (ooooo)

measurements made near Toulouse, the nuclei content as deduced from the small ion content presents better resolution than a direct measurement. The difference in the time constants for the two methods (one second for small ions, one minute for nuclei) introduces more marked deviations between concentrations than in the preceding study. We note that, in this case, the width of the polluted zone is less (6 km), and that the concentrations in the plume are highly variable. Particle flux determinations for each of the two detection methods are, respectively, $7.0 \cdot 10^{17} \pm 1.9 \cdot 10^{17}$ (indirect method), and $6.1 \cdot 10^{17} \pm 1.6 \cdot 10^{17}$ (direct method) particles s^{-1}. Differences observed between these two values are within the margins of error.

The utilization of small ions permits a study with better spatial resolution of the repartition of concentrations.

In Table 3, we give flux values for each measurement.

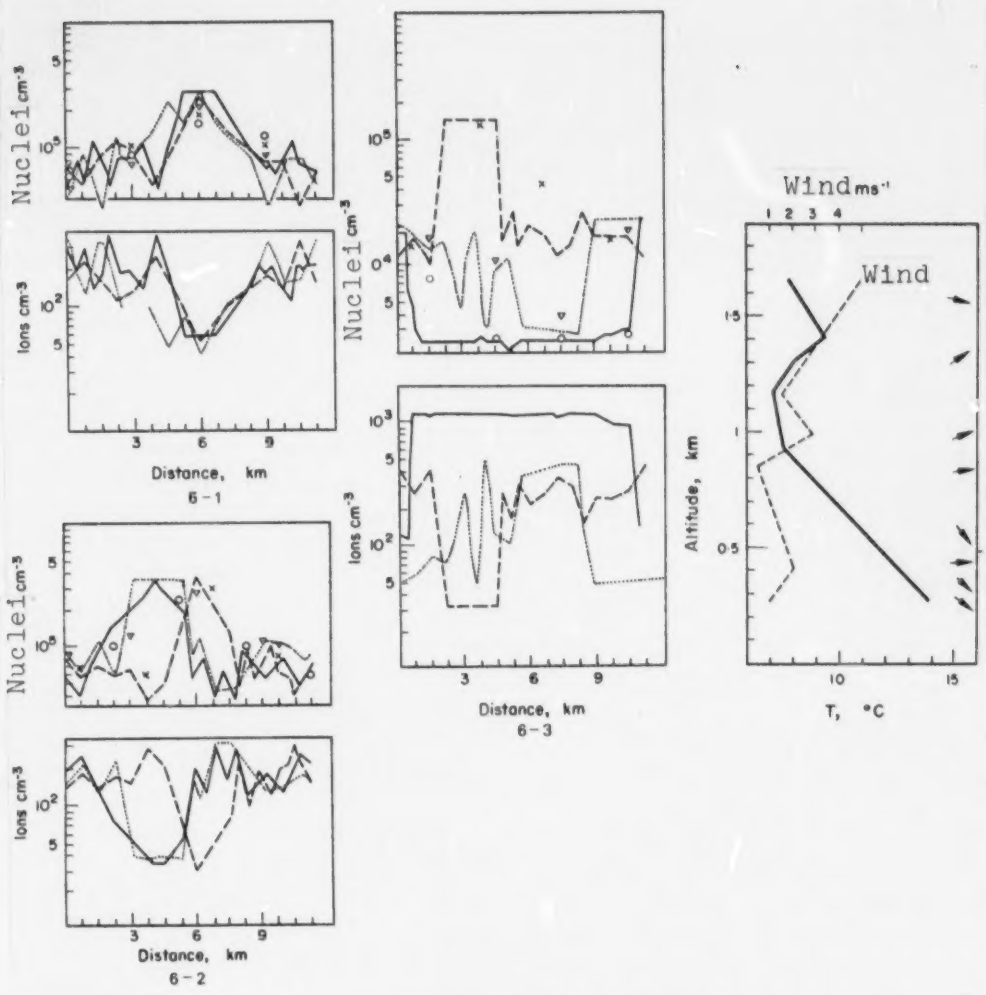

Figure 6. Experiment on September 13, 1972, in the plume from the Lacq industrial complex at 5 km from the center of emission. 6 - 1: measurement level for small ion concentration and calculated nuclei concentration values: 150 m (---); 225 m (····); 300 m (——); direct measurement of nuclei concentration: 150 m (×); 225 m (▽); 300 m (o). 6 - 2 measurement level for small ion concentration and calculated nuclei concentration values: 450 m (----); 600 m (····); 750 m (——); direct measurement of nuclei concentration: 450 m (×); 600 m (▽); 750 m (o). 6 - 3: measurement level for small ion concentration and calculated nuclei concentration values: 900 m (----); 1050 m (····); 1200 m (——); direct measurement of nuclei concentration: 900 m (×); 1050 m (▽); 1200 m (o)

TABLE 3. FLUX VALUES FOR AIKEN NUCLEI EMITTED BY THE LACQ INDUSTRIAL COMPLEX

Date and hour	Distance from city (km)	Polluted zone width (km)	Mean wind speed (ms^{-1})	Particle flux (particles s^{-1})	Observations
June 21, 1972 10:00 h	4	9	2	$8{,}5.10^{17} \pm 2{,}6.10^{17}$	Plume limited in altitude by temperature inversion at 900 m
June 21, 1972	15	20	2	$2{,}0.10^{18} \pm 6{,}8.10^{17}$	Conditions identical to those preceding
Sept. 13, 1972 15:00 h	4	6	1	$7{,}0.10^{17} \pm 1{,}9.10^{17}$	Plume limited in altitude by temperature inversion situated at 1200 m

Flux values near $1 \cdot 10^{18}$ particles s^{-1} are determined with 27 - 34% accuracy. In this case, with concentrations generally greater than $1 \cdot 10^{5}$ particles cm^{-3}, the principal source of error lies in the measurements of space and time parameters given by the airplane.

1.4. Determination of the rate of nuclei formation in the plume of a source. The total quantity of nuclei (C) contained in a vertical section of thickness Δp varies as a function of the distance from the source. In certain cases, there may occur nuclei formations within the polluted zone; in all cases, there are disappearances linked with coagulation.

To take these phenomena into account, we may write the equation:

$$\frac{dC}{dx} = \frac{1}{V}(\tau_1 - \text{loss}). \qquad (7)$$

Supposing the particle concentration in plume thickness Δ_p to be constant, we write:

$$C = C_T . \Delta_p$$

C_T — total quantity of nuclei from the pollution source within a 1 cm thickness of the plume. At distance x_1 from the source, this may be expressed:

$$C_{T_{x_1}} = \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} C_{x_1}(y,z) - C_{x_{1,0}}(z)\, dy\, dz.$$

C_{x_1} and $C_{x_{1,0}}$ — respectively, the volume concentrations inside and outside the polluted zone;

V — wind speed;

τ_1 — total quantity of nuclei formed per second in the plume thickness Δp.

It is likely that nuclei form in the plume as a result of transformation reactions of gaseous constituents emitted by the source. Their rate of formation must, therefore, evolve with the durations of these gases in the polluted atmosphere, admittedly more than several hours.

We have studied the evolution of polluted zones for the urban district of Toulouse and the industrial complex of Lacq at distances of less than 30 km from the source, and at time intervals of less than 2 hours (distance between two vertical measurement sections less than 20 km). We will consider in these conditions that the rate of formation (τ_1) remains constant as a function of distance from the source in the volume of the polluted zone delimited by the two vertical measurement sections.

Moreover, the total quantity of nuclei formed per second in a plume thickness Δp is characteristic of the pollution source, varying from one experiment to another with the wind speed. In order to rid ourselves of this variation, we consider a plume thickness Δp such that $\Delta p/V = 1$.

1.1.2. Evaluation of loss. Losses due to coagulation alter volume concentrations of nuclei in the plume. In particles per second, the rate of disappearance in a vertical section of the

polluted zone of thickness Δp may be written:

$$\int_{-\infty}^{+\infty}\int_{-\infty}^{+\infty} kC^2_{(x,y,z)}\,\Delta p\,.\,dy\,dz$$

where k — a coagulation constant equal to 10^{-9} cm^3 s^{-1} for particles of mean radius $3 \cdot 10^{-6}$ cm.

As we have mentioned, the limits of integration are, respectively, in directions y and z, the width and height of the plume. These limits vary as a function of distance from the source, due to diffusion phenomena. Strictly speaking, an estimate of the loss due to coagulation will be obtained only through the resolution of a diffusion equation into which the coagulation term has been introduced. This is not the object of our study.

We suppose, to a first approximation, that the disappearance of nuclei within the polluted zone is proportional to the square of the mean concentration in the volume delimited by the two vertical sections of the plume at distances x_1 and x_2 from the point of emission. We liken this volume to a cylinder of vertical section $\overline{S}$, and we further suppose that the evolution of the plume between x_1 and x_2 proceeds according to a linear law of variation. Under these conditions, we may express the disappearance as:

$$k\,.\,\overline{C}^2\,\overline{S}\Delta p. \qquad (8)$$

$\overline{C}$ — mean volume concentration in the plume, equal to:

$$\frac{\overline{C_{M_{x_1}}} + \overline{C_{M_{x_2}}}}{2}$$

$\overline{C_{M_{x_1}}}, \overline{C_{M_{x_2}}}$ — mean volume concentrations in vertical measurement sections at distances x_1 and x_2 from the source;

$\overline{S}$ — mean area of vertical plume sections, equal to $(S_{x_1} + S_{x_2})/2$;

S_{x_1}, S_{x_2} — areas of sections of the polluted zone at distances x_1 and x_2 from the source.

1.1.3. Estimation of formation rate in the plume. In more explicit terms, Equation (7) may be represented:

$$\Delta p \frac{dC_T}{dx} = \frac{1}{V}(\tau_1 - k\overline{C}^2\overline{S}\,\Delta p).$$

Its integration between x_1 and x_2 gives:

$$(\tau_1)_{x_1}^{x_2} = \left\{ \frac{V(C_{T_{x_2}} - C_{T_{x_1}} + k\overline{C}^2\overline{S}(x_2 - x_1))}{x_2 - x_1} \right\} \Delta p$$

or:

$$(\tau_1)_{x_1}^{x_2} = \left\{ \frac{(\phi_{x_2} - \phi_{x_1}) + k\overline{C}^2\overline{S}(x_2 - x_1))}{x_2 - x_1} \right\} \Delta p. \qquad (9)$$

ϕ_{x_2} and ϕ_{x_1} are the particle flux values through the vertical plume sections at distances x_1 and x_2 from the point of emission.

The accuracy of a nuclei formation rate inside the plume will be closely linked to that of the particle flux values determined at different distances from the source.

An estimate of the error, obtained with the aid of a classical calculation, is given by the expression

$$\frac{\Delta\tau_1}{\tau_1} = \frac{\Delta\phi_{x_1} + \Delta\phi_{x_2} + (\Delta\overline{S})(\overline{kC^2})(x_2 - x_1) + (x_2 - x_1)\overline{S}k\,\Delta\overline{C}^2}{\phi_{x_2} - \phi_{x_1} + k\overline{C}^2\overline{S}(x_2 - x_1)}$$
$$+ \left\{ \frac{\phi_{x_2} - \phi_{x_1}}{(x_2 - x_1)(\phi_{x_2} - \phi_{x_1} + k\overline{C}^2\overline{S}(x_2 - x_1))} \right\} \Delta(x_2 - x_1) + \frac{\Delta(\Delta p)}{\Delta p}. \qquad (10)$$

We estimate the relative error in distance separating the two vertical sections at 10 percent. Error for Δp is the same as that obtained for wind intensity measurements (on the order of 15%).

Experimental results in the urban district of Toulouse and in the Lacq industrial complex

a) Urban Toulouse. At 10 and 30 km from the center, two profiles, made on April 25, 1972 at an interval of one hour, give respective flux values of $5.8 \cdot 10^{17} \pm 2.9 \cdot 10^{17}$ and

$5.4 \cdot 10^{17} \pm 2.7 \cdot 10^{17}$ particles s^{-1}. These values are of the same order of magnitude. They show that the rate of nuclei formation in the plume is low; the margins of error (about 50 percent) will not allow us to assign it any significant value.

b) Lacq industrial complex. Two measurements made on June 21, 1972, indicate an augmentation in flux with increasing distance from the source ($8.5 \cdot 10^{17} \pm 2.6 \cdot 10^{17}$ particles s^{-1} at 4 km, $2 \cdot 10^{18} \pm 6.8 \cdot 10^{17}$ particles s^{-1} at 15 km), evidencing the formation of nuclei in the plume. The wind speed remained constant: 2 ± 0.3 ms^{-1}. The width of the polluted zone increased from 9 to 20 km. The height, limited by a temperature inversion at 900 m, did not vary. The mean concentration in the volume delimited by the two vertical sections is $7.8 \cdot 10^{4} \pm 1.2 \cdot 10^{4}$ particles cm^{-3}.

These values indicate a nuclei source intensity, in a portion of the plume of thickness $\Delta p = 2$ m, and covering the distance between 4 and 15 km from the emission zone, of $3.8 \cdot 10^{14} \pm 2.8 \cdot 10^{14}$ particles s^{-1}.

2. Determination of Formation Rate in the Case of a Source of Large Surface Area — Application to a Natural Source: The Landes Forest

2.1. Method. This consists in comparing the total quantities of nuclei contained in 1 cm^2 columns of atmosphere. These are obtained by means of vertical repartitions executed above the source at different distances, in the direction of the wind.

Supposing that the nuclei concentration is uniform in the mix stratum, which has been verified in general during the day (Lopez et al., 1974), the equation for evolution of nuclei may be written:

$$\frac{dC}{dx} = \frac{1}{V}\left(\frac{\tau_1}{h} - kC^2\right). \qquad (11)$$

C — volume concentration in the mix stratum;

τ_1 — formation rate in this stratum;

h — height of the mix stratum;

k — coagulation constant, equal to 10^{-9} cm^3 s^{-1}, for particles of mean radius $3 \cdot 10^{-6}$ cm;

V — wind speed.

This equation was resolved in a preceding work (Lopez et al., 1973).

The accuracy of this method may be estimated to a first approximation, neglecting coagulation phenomena, by the expression:

$$\frac{\Delta\tau_1}{\tau_1} = \frac{\Delta V}{V} + \frac{\Delta h}{h} + \frac{\Delta C_2}{C_2 - C_1} + \frac{\Delta C_1}{C_2 - C_1} + \frac{\Delta(\Delta x)}{\Delta x}.$$

Δx — distance between two vertical repartitions.

Replacing the different terms by their values, this expression becomes:

$$\frac{\Delta\tau_1}{\tau_1} = 30\% + \frac{\Delta C_2}{C_2 - C_1} + \frac{\Delta C_1}{C_2 - C_1}. \qquad (12)$$

C_1 and C_2 represent volume concentrations in the mix stratum for the two profiles under consideration.

2.2. Application to the Landes forest. The forested zone of Landes borders on the Atlantic coast, and covers an area of 7000 km^2. The vegetation is coniferous.

In Figure 7 we present 4 vertical profiles of the nuclei concentrations executed between Toulouse and the Atlantic coast in the direction of the wind (southwest). In Landes profiles 3 and 4, we note an augmentation in the height of the mix stratum, linked with the thermal profile. We also note an increase in the total quantity of nuclei in a 1 cm^2 column (profile 3: $9.2 \cdot 10^8$ particles; profile

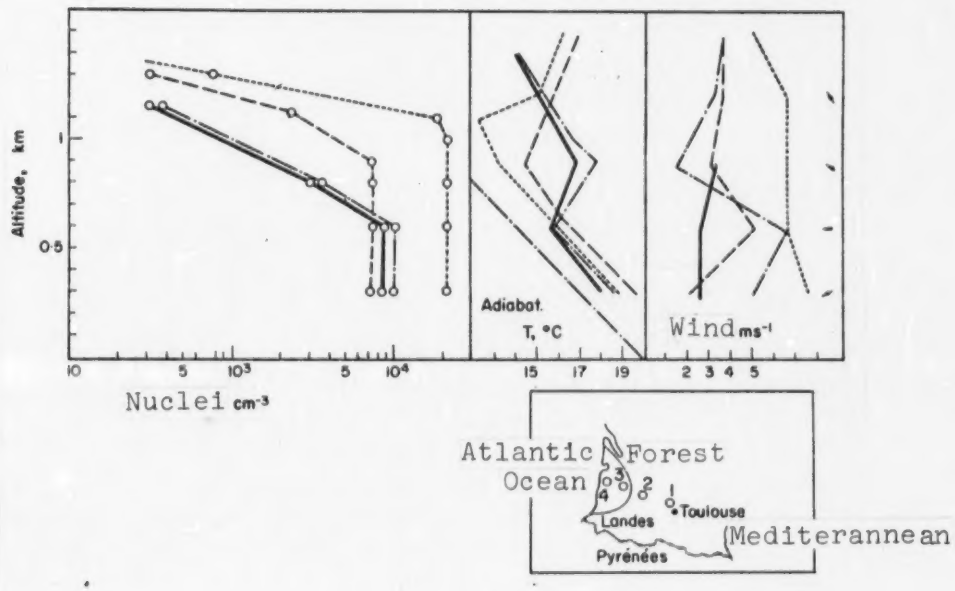

Figure 7. Experiment on June 24, 1972. Evolution of the vertical repartition of Aitken nuclei between Toulouse and the Atlantic coast, running southwest. Profile 1 (——); profile 2 (-.-.-); profile 3 (----); profile 4 (····)

4: $2.5 \cdot 10^9$ particles), while outside the forested zone, this quantity is practically constant (profile 1: $8.1 \cdot 10^8$ particles; profile 2: $8.5 \cdot 10^8$ particles). This increase indicates a nuclei source of greater intensity above the forested region.

The length of time which the air mass remained above the forest in profiles 3 and 4 is on the order of 4 hours for a mean wind speed 5 ms^{-1}. The volume concentration of nuclei in the mix stratum increases during this time from 8000 to 20,000 particles cm^{-3}.

Under such conditions, we obtain a mean formation rate in the forested zone of $1.2 \cdot 10^5 \pm 5.3 \cdot 10^4$ particles cm^{-2} s^{-1}.

This value is obviously 3 times greater than the mean formation rate determined in a rural zone of southwestern France (Lopez et al.,

1973); it is in good agreement with the value determined in a preceding study ($1 \cdot 10^5$ particles cm^{-2} s^{-1}) by comparing vertical repartitions of Aitken nuclei and natural radioactive tracers, lead-212 and radon-222 at different distances from the coast (Lopez et al., 1974).

3. Method for Determination of Formation Rate in the Case of a Uniformly Distributed Source

3.1. Method. This consists of comparing the total quantities of nuclei contained in 1 cm^2 columns of air, measured at different times during the day, in the same sample zone and under identical wind conditions.

The equation for nuclei evolution as a function of time will be written, as in the preceding case, by supposing a homogeneous repartition in the mix stratum:

$$\frac{dC}{dt} = \frac{\tau_1}{h} - kC^2 \qquad (13)$$

The different terms in this expression have been explained (see 2.1). Its resolution is given in a previously cited work. The accuracy of τ_1 may be estimated, to a first approximation, neglecting coagulation phenomena, by the expression:

$$\frac{\Delta\tau_1}{\tau_1} = \frac{\Delta h}{h} + \frac{\Delta C_{(t_1)}}{C_{(t_2)} - C_{(t_1)}} + \frac{\Delta C_{(t_2)}}{C_{(t_2)} - C_{(t_1)}}. \qquad (14)$$

$C(t_1)$ and $C(t_2)$ represent, respectively, the volume concentrations in the mix stratum measured at instants t_1 and t_2.

3.2. Application to the determination of the mean nuclei formation rate in the mix stratum of a non-polluted zone. We have shown in a preceding study (Lopez et al., 1974) that outside zones of significant pollution, the source of Aitken nuclei, in the mix stratum, may be considered as relatively well distributed along the path of an air mass.

The formation rate of nuclei in this stratum may be determined, according to the method presented in the above paragraphs, from vertical profiles graduated according to time of day.

Characteristics of the site have been described in a preceding article (Lopez et al., 1974). *. . . rural in the Toulouse region in anti-cyclonic periods and in the absence of fog.

(a) Evaluation of mean formation rate during the course of the day

Case where a single profile was executed at the end of the day: In certain cases, we made only one profile at the end of the day (between 16:00 and 17:00 hours). We suppose, in these experiments, that the measured nuclei concentration is an equilibrium value, that is to say, $dC/dt = 0$.

Under such conditions, the formation rate is expressed by:

$$\tau_1 = khC_E^2.$$

C_E — volume concentration in the mix stratum at equilibrium.

One may estimate, in the lower strata of the atmosphere where the pseudo period of the nuclei is between 10 and 20 hours, and where the source intensity varies according to a diurnal cycle (Lopez et al., 1973 - 1974), that the error introduced by this hypothesis is less than a factor of 2. Values thus obtained are given in Table 4. The mean intensity of the diurnal source is between $1.9 \cdot 10^4$ and $3.8 \cdot 10^4$ particles cm^{-2} s^{-1}. These values confirm those obtained from measurements executed at ground level.

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*Translator's note. There is a printer's omission here in the original foreign text. The first sentence of the paragraph is repeated following the end of the second (incomplete) sentence.

Variation in the formation rate during the course of the day: the formation rate of nuclei may be determined at different times of the day from vertical profiles graduated in time.

In this case, the initial concentration necessary for resolution of Equation (13) is that obtained on the preceding flight.

TABLE 4. MEAN DIURNAL FORMATION RATES FOR AITKEN NUCLEI IN THE MIX STRATUM

Day	h (m)	τ_1 ($p\,cm^{-2}\,s^{-1}$)	τ_1/h ($p\,cm^{-3}\,s^{-1}$)
September 17, 1971	800	$1{,}9.10^4$–$3{,}8.10^4$	0,2–0,5
September 29, 1971	700	$2{,}8.10^4$–$5{,}6.10^4$	0,4–0,8
March 17, 1972	1000	$3{,}3.10^4$–$6{,}6.10^4$	0,3–0,7
May 19, 1972	1500	$4{,}2.10^4$–$8{,}5.10^4$	0,3–0,6
May 29, 1972	1000	$1{,}7.10^4$–$3{,}4.10^4$	0,2–0,3
May 31, 1972	1600	$1{,}6.10^4$–$3{,}2.10^4$	0,1–0,2
September 8, 1972	1100	$8{,}8.10^3$–$1{,}8.10^4$	0,1–0,2
September 9, 1972	1400	$1{,}4.10^4$–$2{,}8.10^4$	0,1–0,2
September 10, 1972	1700	$8{,}3.10^3$–$1{,}7.10^4$	0,05–0,1
September 11, 1972	1600	$9{,}1.10^3$–$1{,}8.10^4$	0,06–0,1
September 12, 1972	1300	$2{,}8.10^4$–$5{,}6.10^4$	0,2–0,4
September 13, 1972	1500	$1{,}5.10^4$–$3{,}0.10^4$	0,1–0,2
September 16, 1971	1400	$1{,}1.10^4$–$2{,}3.10^4$	0,1–0,2
October 15, 1971	850	$1{,}7.10^4$–$3{,}4.10^4$	0,2–0,4
September 27, 1971	1000	$2{,}3.10^4$–$4{,}6.10^4$	0,2–0,5
Mean values	1200	$1{,}9.10^4$–$3{,}8.10^4$	0,2–0,3

h — height of the mix stratum.
τ_1 — formation rate in a 1 cm^2 column of atmosphere (particles $cm^{-2}\ s^{-1}$).
τ_1/h — volume formation rate (particles $cm^{-3}\ s^{-1}$).

We present as examples (Figure 8 - 11) several of these vertical repartitions obtained at different times during the day. A detailed analysis of these profiles will follow.

Results thus obtained are given in Table 5. We note that the formation rate remains practically constant during the course of the day, and of the same order of magnitude as the mean daily value (Table 4). If we consider a uniform formation in the mix stratum, then the volume intensity of the nuclei source varies during the course of the day; in the inversion stratum, which is situated near

TABLE 5. VARIATION IN THE FORMATION RATE OF THE AITKEN NUCLEI DURING THE COURSE OF A DAY IN THE MIX STRATUM ABOVE THE TOULOUSE REGION

Day	h	$\tau_1 (p\,cm^{-3}\,s^{-1})$	$\tau_1/h\,(p\,cm^{-3}\,s^{-1})$
October 15, 1971	sunrise-10h*	$2.6.10^4 \pm 1.0.10^4$	1.2 ± 0.5
	10:00-15:00	$3.10^4 \pm 1.6.10^4$	0.35 ± 0.2
	15:00-17:00	$3.10^4 \pm 2.10^4$	0.35 ± 0.23
September 27, 1971	sunrise* 10:00	$2.5.10^4 \pm 1.0.10^4$	1.0 ± 0.4
	10:00-16:00	$3.1.10^4 \pm 1.2.10^4$	0.3 ± 0.1
November 17, 1971	sunrise* 11:00	$7.0.10^4 \pm 2.2.10^4$	1.4 ± 0.4
September 9, 1972	sunrise* 10:00	$3.7.10^4 \pm 1.5.10^4$	1.5 ± 0.6
September 13, 1972	sunrise* 10:00	$5.6.10^4 \pm 1.9.10^4$	1.4 ± 0.5

*At sunrise, we take the concentration measured above the station as our initial value.
τ1/h — represents the volume formation rate in the mix stratum of height h, supposing that the intensity of source y is uniform.

the ground in the morning, it is between 1 and 1.4 particles $cm^{-3*}\ s^{-1}$, a value 5 to 10 times higher than the mean daily value of 0.17 to 0.34 particles $cm^{-3}\ s^{-1}$.

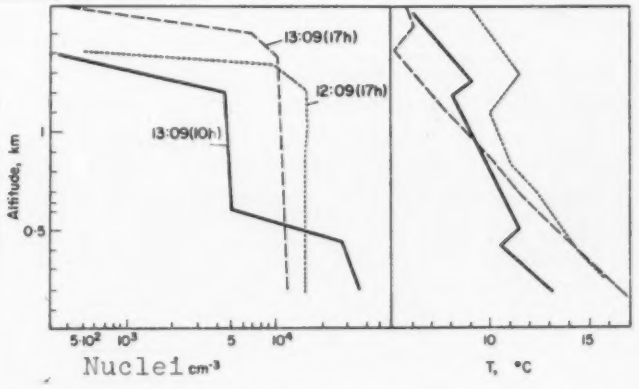

Figure 8. Experiment on September 12 and 13, 1972. Vertical profile of Aitken nuclei: September 12, 1972 at 17:00 h (·····); September 13, 1972 at 10:00 h (——); September 13, 1972 at 17:00 H (----)

(b) Interpretation of vertical profiles obtained at different times during the day

At 10:00 in the morning (Figures 8 - 10), we note strong concentrations of nuclei on the order of 10:00 (Lopez, 1974) . . .** the concentrations observed on September 13, that they are lower above (2000 - 5000 particles cm-3).

*Translator's note. Erroneously printed in foreign text as cm^3.
**Translator's note. Printer's omission occurs here in foreign text.

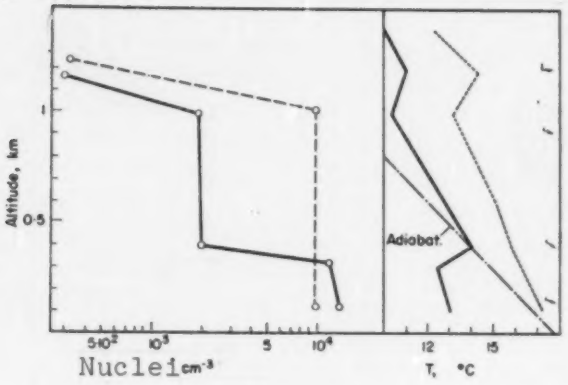

Figure 9. Experiment on September 27, 1971. Vertical repartition of Aitken nuclei above the Toulouse region at 10:00 h (——); at 17:00 H (-----)

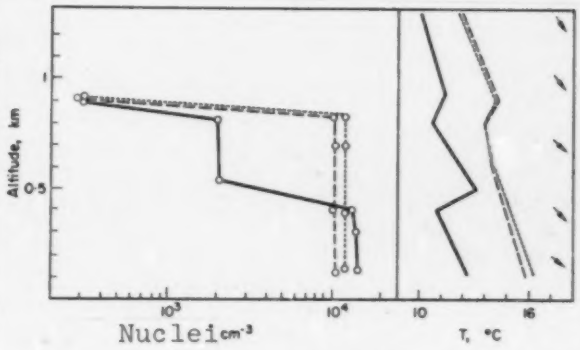

Figure 10. Experiment on November 17, 1971. Vertical repartition of Aitken nuclei above the Toulouse region at 10:00 h (——); at 15:00 h (----); at 17:00 h (.....)

These latter values are essentially the same as those observed at 5:00 in the morning for different days (Figure 11, for example). The development of this situation is illustrated in Figure 8. On September 12, 1972, at 17:00 hours, we note a homogeneous concentration in the mix stratum (0 - 1300 m). A sharp gradient at 1300 m is due to the presence of a temperature inversion, inhibiting exchanges between this stratum and the free atmosphere.

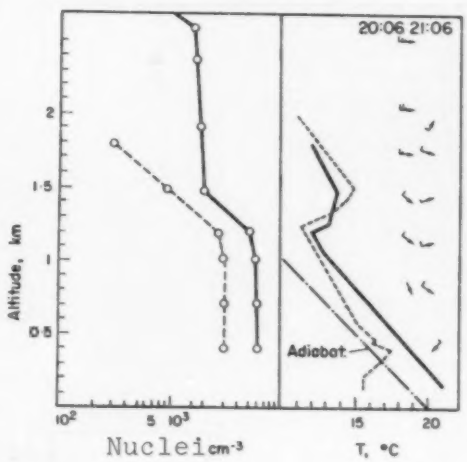

Figure 11. Experiment on June 20 and 21, 1972. Vertical profile of Aitken nuclei: June 20 at 17:00 h (——); and June 21, 1972 at 05:00 h (---)

During the night, this temperature inversion is maintained, while a stable stratum develops near the ground to a height of 400 m, limiting exchange of materials arising from the soil.

In this 400 - 1300 m stratum, the nocturnal decrease in concentration is thus due principally, in the absence of exchanges with

higher and lower strata, to coagulation phenomena (meteorological analysis indicates absence of any lixiviation during the night).

Considering a nocturnal pseudo period of nuclei disappearance in this stratum on the order of 10:00 h (Lopez, 1974), concentrations observed September 13, 1972 at 10:00 would indicate that the formation rate in this stratum during the night and after sunrise until 10:00 a.m. remains low.

During the course of the day, the temperature inversion near the ground disappears. We observe an augmentation in the total quantity of nuclei in the atmosphere, accompanied by a homogenization of concentrations.

These different observations suggest two complementary hypotheses for the diurnal formation of nuclei in the mix stratum.

In the nocturnal inversion stratum, there would occur: either a direct formation of nuclei, or sunlight-related transformation reactions of gases emitted by the soil or vegetation. The latter hypothesis appears more likely when results obtained at ground level are taken into account (Lopez et al., 1973). This would imply continuous emission as a function of time of one or several of the constituents participating in these transformation reactions.

Above the nocturnal inversion stratum, little nuclei formation occurs. This indicates that the lifespan of one or several of the constituents participating in the gas-solid and gas-liquid transformations (the sole origin of higher-altitude nuclei formation) is less than 12 hours.

These results specifically confirm earlier findings (Lopez et al., 1974), which enabled us to show that the production rate above the temperature inversion layer (near the ground at night, at the upper limit of the mix stratum during the day) is low.

4. Balance of Aitken Nuclei Sources in the Southwestern Region of France

The Midi-Pyrenees and Aquitaine regions, in which these measurements were made, have a surface area of 87,000 km^2, and a population of 4 million. They present diverse and highly localized nuclei sources (urban districts, industrial complexes, natural sources).

Intensities of several of these principal sources were determined with the aid of methods exposed in this article. Through a first balance of our results, we are able to estimate the relative importance of natural and artificial sources in this region.

The urban district of Toulouse, population 400,000, generates, on the average, with its industrial environment, $5 \cdot 10^{17}$ particles s^{-1}. This value corresponds to an emission of $1.25 \cdot 10^{12}$ particles s^{-1} $person^{-1}$. For the group of regions under consideration, the implied flux is $5 \cdot 10^{18}$ particles s^{-1}, if we assume that the industrial environment of the Toulouse urban district is representative of others in the region being studied.

Studies executed both on the ground and in the air in rural zones give a diurnal nuclei formation rate of $3 \cdot 10^{4}$ particles cm^{-2} s^{-1}, or, for the entire region, $2.6 \cdot 10^{19}$ particles s^{-1}.

Comparison of the preceding figures shows that 25 percent of the nuclei are of artificial origin, and 75 percent are of natural origin. These values, which constitute only the order of magnitude, might be determined with greater precision through complementary measurements, using methods exposed previously.

The Landes forest, with its essentially coniferous vegetation, is an important natural source covering 7000 km^2. The intensity of this source, measured in the months of May and June, is on the order

of 10^5 particles cm^{-2} s^{-1} or for the entire zone, 7×10^{18} particles s^{-1}. If we consider, according to Went (1966), that these nuclei result from photo-oxidation reactions of gaseous constituents emanating from the vegetation (α, β-pinene, myrcene, isoprene ...), it is probable that nuclei will also form beyond the forested zone. The distance over which the influence of this source is felt must depend on the duration of these gases in the atmosphere, about which very little is known. We may thus assume that the Landes forest, which, according to measurements made above the forested zone, generates 25 percent of the nuclei of the Midi-Pyrenees and Acquitaine regions, is in fact an even more important source.

Among the industrial sources, one of the most important is the Lacq complex, with a flux at several km from the source on the order of $1 \cdot 10^{18}$ particles s^{-1}, or approximately 4 percent of the total quantity of nuclei generated by the region. As in the preceding case, this value is deficient, in view of the fact that there occurs a continuous production of nuclei in the polluted zone (about $1.5 \cdot 10^{12}$ particles cm^{-2} s^{-1} near the source). The importance of this production, situated outside the industrial zone, will depend on the formation mechanisms of the nuclei and on the lifespans of the polluting gases that participate in these reactions.

Conclusion

With the aid of aerial measurements, we have developed different methods for determination of the intensities of Aitken nuclei sources in the atmosphere. This has permitted the study of several of the principal nuclei sources in southwestern France.

These sources may be classed into two groups: point-sources, which are generally the urban districts and the industrial complexes, and extended sources, which generally include zones of countryside and forest.

For the first group, the method utilized consists of determining the particle flux through a vertical section of the plume. This is measured through a spatial repartitioning of particle concentrations in the polluted zone.

Concentrations are obtained either 1) directly, by use of a nuclei counter for which the time constant is about one minute, or 2) indirectly, by use of electrostatic precipitators that give the small ion content, and for which the time constant is on the order of a second.

In our measurements, these two sampling methods have given similar values for each of the sources studied (urban district of Toulouse, Lacq industrial complex). The use of small ions allows us to obtain a better spatial resolution of the concentration repartition within the polluted zone. It will also permit further study of sources with smaller geometric dimensions, for which the time constant of the nuclei counter would introduce significant error.

The accuracy of this method varies with the dilution. For example, the error is between 35 and 50 percent in the case of an urban district of population 400,000. Its utilization will thus be limited in the case of smaller urban districts.

For the second group, we studied the evolution of the vertical repartition of concentrations as a function of time or of space. The corresponding accuracy is a function of source intensity.

In the particular case where the population density is 46 inhabitants km^{-2}, our first balance shows that natural sources are more important than artificial sources (75 percent compared with 25 percent). Among the natural sources, we note that the forested zones of Landes, which is covered with essentially coniferous vegetation, furnishes, at least during daytime in spring, three times more nuclei per unit of surface and time than the open countryside ($1 \cdot 10^5$ particles cm^{-2} s^{-1}, compared to $0.3 \cdot 10^5$ particles cm^{-2} s^{-1}).